Field-Induced Quantum Criticality and Universal Temperature Dependence of the Magnetization of a Spin-1/2 Heisenberg Chain

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High-precision dc magnetization measurements have been made on $Cu(C_4H_4N_2)(NO_3)_2$ in magnetic fields up to 14.7 T, slightly above the saturation field $H_s = 13.97$ T, in the temperature range from 0.08 to 15 K. The magnetization curve and differential susceptibility at the lowest temperature show excellent agreement with exact theoretical results for the spin-1/2 Heisenberg antiferromagnet in one dimension. A broad peak is observed in magnetization measured as a function of temperature, signaling a crossover to a low-temperature Tomonaga-Luttinger-liquid regime. With an increasing field, the peak moves gradually to lower temperatures, compressing the regime, and, at H_s, the magnetization exhibits a strong upturn. This quantum critical behavior of the magnetization and that of the specific heat withstand quantitative tests against theory, demonstrating that the material is a practically perfect one-dimensional spin-1/2 Heisenberg antiferromagnet.

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Quantum spin systems in one dimension have been the subject of intensive experimental and theoretical studies because of their intriguing properties arising from strong quantum fluctuations [1]. Among them, one of the simplest is the spin-1/2 one-dimensional (1D) Heisenberg antiferromagnet (HAF), whose ground state is a quantum critical state called a Tomonaga-Luttinger liquid (TLL) [2]. Two hallmarks of this unique state are gapless elementary excitations, which are interacting spin-1/2 quasiparticles known as spinons, and power-law decays of correlation functions indicating a quasi-long-range order [1]. The basic character of the TLL in this system has been well established theoretically, yet quantitative comparisons with experiment are still incomplete, particularly near the saturation magnetic field.

In a magnetic field H, the Hamiltonian of the spin-1/2 1D HAF is

$$\mathcal{H} = J\sum_i \boldsymbol{S_i} \cdot \boldsymbol{S_{i+1}} - g\mu_B H \sum_i S_{iz}, \tag{1}$$

where J is the intrachain coupling constant, and g and μ_B are the g factor and the Bohr magneton, respectively. The TLL survives up to the saturation field $H_s = 2J/g\mu_B$ [1–3], the quantum critical point (QCP)—in fact, the end point of a line of quantum critical points—at which it gives way to a gapped, field-induced ferromagnetic state.

In 1D spin systems that are gapped at zero field, such as spin-1 Haldane chains and spin-1/2 two-leg ladders, an additional QCP exists—the lower critical field H_c, at which a quantum phase transition takes place from a gapped, disordered state to a TLL. Near H_s and H_c, an effective description of the TLL is given in terms of interacting magnons—quasiparticles carrying spin 1 [4,5]; the ground states in the regions $H \geq H_s$ and $H \leq H_c$ can be considered vacuums, in which excitations are, respectively, $S_z = -1$ and $S_z = 1$ magnons [6].

In the dilute limit, these 1D magnons can be exactly mapped onto free fermions [7,8]. As a result, the number of magnons, N_m, near the QCPs is given by

$$\begin{aligned}\frac{N_m}{L} &= \int_0^\infty d\epsilon D(\epsilon) f(\epsilon - \mu) \\ &= \frac{\sqrt{2mk_BT}}{\pi\hbar} \int_0^\infty \frac{dx}{e^{x^2-\mu/k_BT}+1},\end{aligned} \tag{2}$$

where L is the number of spins, $f(\epsilon-\mu)$ the Fermi distribution function, and $D(\epsilon)$ the density of states of the free fermions, whose dispersion at the band edge is quadratic, $\epsilon = \hbar^2k^2/2m$. Here, m is the effective mass, and the chemical potential μ is $g\mu_B(H_s - H)$ or $g\mu_B(H - H_c)$ [6,9]. Magnetization per spin, M/L, is $(M_s - N_m)/L$ and N_m/L near H_s and H_c, respectively, where M_s is the saturation magnetization.

According to Eq. (2), the magnetization at a given μ has an extremum at [6]

$$k_BT_{\rm ex} = 0.76238\mu, \tag{3}$$

where N_m becomes minimum. This universal relation, confirmed in the spin-ladder system $(Cu_7H_{10}N)_2CuBr_2$ (DIMPY) near $H_c = 3$ T [10], marks the boundary at which the quadratic dispersion becomes a poor approximation because of the linear dispersion of spinons near $\varepsilon = 0$—a crossover from a quantum-critical region to a TLL region [6,9,11]. The magnetization extremum persists even at fields far away from the QCPs, as has been shown by numerical calculations for spin-1 Haldane chains [6] and spin-1/2 two-leg spin ladders [12,13], and as has been observed in DIMPY [10], $Ni(C_5H_{14}N_2)_2N_3(PF_6)$ (NDMAP) [14], and $(Cu_5H_{12}N)_2CuBr_4$ (BPCB) [15]. This easily identifiable anomaly in magnetization, hence, serves as a convenient marker of the crossover to the low-temperature, TLL region at all fields [15]. However, in spin-1/2 1D HAFs, no experimental work has been done to our knowledge to investigate a temperature dependence of magnetization in detail at fields near H_s, the QCP, because many spin-1/2 1D HAFs, including Sr_2CuO_3 ($J = 2200$ K) [16] and $KCuF_3$ ($J = 380$ K) [17], need very strong magnetic fields, in excess of hundreds of teslas, to reach H_s.

In this Letter, we investigate quantum critical behavior of the magnetization of a spin-1/2 1D HAF near this QCP in detail. For this purpose, we have performed high-precision dc magnetization measurements, supplemented by some specific-heat measurements, on $Cu(C_4H_4N_2)(NO_3)_2$, or CuPzN for short—a prototypical spin-1/2 1D HAF compound with a relatively small intrachain coupling of $J = 10.3$ K [18] and a corresponding H_s of about 14 T. Comparison of the magnetization data, taken at 0.08 K which is less than $0.01J$, with a Bethe-ansatz prediction and our exact calculation employing the quantum transfer-matrix (QTM) method [19,20] demonstrates that CuPzN is a practically perfect spin-1/2 1D HAF. We observe quantum critical behavior near the QCP in excellent agreement with Eqs. (2) and (3) and with QTM results. Preliminary results have been reported in Ref. [21].

In CuPzN, chains of $S = 1/2$ Cu^{2+} run along the crystallographic ***a*** axis [18,22]. A zero-field muon-spin-relaxation experiment has detected three-dimensional (3D) magnetic ordering at $T_N = 0.107$ K [23]. From this, the interchain coupling constant J' has been estimated to be 0.046 K. Consistent with such a small J' relative to J, no anomaly indicative of the ordering has been found in specific heat and magnetization down to 0.05 K, well below T_N [24].

Our dc magnetization measurements were performed on a 3.59 mg sample of CuPzN, using a force magnetometer [25]. A ^{3}He-^{4}He dilution refrigerator and a sorption-type ^{3}He refrigerator were used in the temperature ranges $0.08\ \mathrm{K} \le T \le 2$ K and $0.3\ \mathrm{K} \le T \le 15$ K, respectively. Static magnetic fields up to 14.7 T were applied along the ***b*** axis, perpendicular to the spin-chain direction. Precise calibration of the magnetization was made by comparing the $M(H)$ data at 4.2 K with those obtained by a superconducting-quantum-interference-device magnetometer. In addition, specific-heat measurements were performed on a 1.10 mg sample at 14 T with a relaxation technique. The samples for both measurements were single crystals grown by slow evaporation of a mixture of deuterated pyrazine with a heavy-water solution of copper nitrate [22].

Figure 1(a) shows the magnetization M and the magnetic susceptibility dM/dH of CuPzN at 0.08 K as a function of the magnetic field up to 14.7 T. Figure 1(b) is an enlarged view of Fig. 1(a) near the saturation field H_s, along with the well-known exact Bethe-ansatz curve at $T = 0$ [26] recomputed for the present purpose. The best fit of the curve to the data gives $J = 10.8(1)$ K and $g = 2.30(1)$, which agree well with previously reported values [18,27], and H_s is found to be 13.97(6) T. The fit is excellent up to 13.9 T, but the data very near H_s do not exhibit a square-root singularity, $M_s - M \propto (H_s - H)^{1/2}$, predicted by theory [28,29]. Accordingly, dM/dH has a prominent peak at 13.95 T but does not diverge. However, fitting the expression $1 - M/M_s = D(1 - H/H_s)^{1/\delta}$ to the data between 13.6 T and 13.9 T yields $D = 1.24(8)$, $H_s = 13.98(1)$ T, and $\delta = 1.98(8)$, with D and δ agreeing with the predicted values $4/\pi \approx 1.273$ [29] and 2, respectively. Moreover, our exact curve for $T = 0.08$ K, calculated by the QTM method and shown in Figs. 1(a) and 1(b), is in close

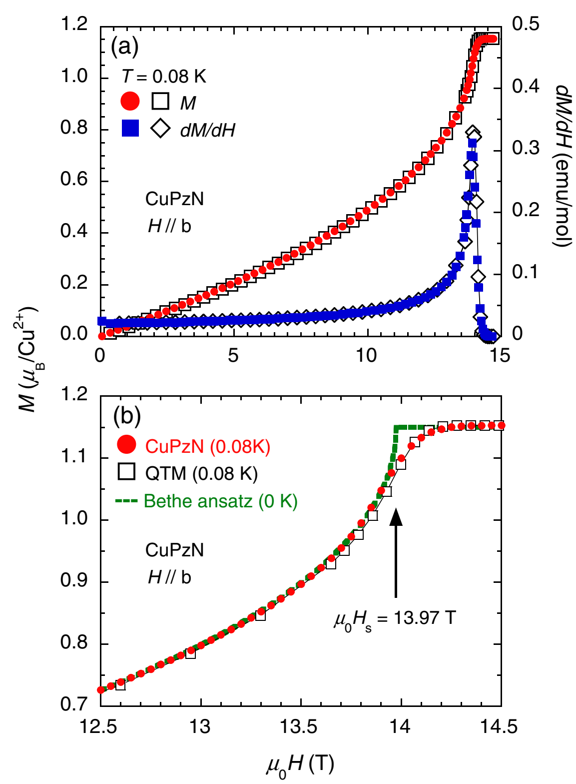

FIG. 1 (color online). (a) Field dependence of the magnetization M (solid circles) and the differential susceptibility dM/dH (solid squares) at 0.08 K, along with the result of exact QTM calculations for the 1D spin-1/2 HAF at 0.08 K (open symbols). (b) Enlarged plot near $H_s = 13.97$ T. The dashed line is a Bethe-anzatz result for $T = 0$. In both panels, thin solid lines are guides to the eye.

agreement with the data even near H_s. These observations strongly suggest that the rounding of M and the corresponding nondivergence of dM/dH at H_s are not caused by the interchain coupling J', but by thermal fluctuations in the vicinity of the QCP [30].

The temperature at which the $M(H)$ curve was measured, 0.08 K, is definitely below the zero-field T_N of 0.107 K. Therefore, the boundary of the 3D ordered phase will cross this temperature at some field below H_s. Nonetheless, the $M(H)$ curve exhibits no anomaly that indicates such a transition, in accordance with the previous experiment on a powder sample [24]. Taken together, these results suggest that the 3D ordering has a negligible effect on the thermodynamic properties of CuPzN.

The temperature dependence of the magnetization is shown in Fig. 2 for several magnetic fields. The magnetization has been divided by the field to compare data taken at different fields. In the limit of $H \to 0$, M/H is expected to reach a maximum at $T_p \sim 0.641J$ [29,31]. This relation, combined with the experimental value of $T_p = 6.89$ K at 1 T, yields $J = 10.8$ K, in perfect agreement with the value determined from the $M(H)$ data. With the increasing field, T_p gradually decreases, and, at 13.9 T, the magnetization peak eventually vanishes into a temperature region well below 0.08 K [see Fig. 2(b)]. At 14 T, the data show a strong upturn as $T \to 0$, indicative of quantum criticality. At fields above H_s, where the ground state is a gapped, field-induced ferromagnetic state, the magnetization levels off at low temperatures as seen in the 14.5 T data. These features have been expected by numerical calculations for spin-1/2 1D HAFs [34].

Figure 3(a) shows the variation of $(M_s - M)/H$ with temperatures for several fields very near H_s in a log-log plot. At 14 T, a field that is indistinguishable from H_s within experimental uncertainty, $(M_s - M)/H$ is approximately proportional to $\sqrt{T}$ down to the lowest temperature investigated; the best fit of the expression $(M_s - M)/H \propto T^{\beta}$ to the data below 1 K yields $\beta = 0.48(1) \approx 1/2$. This power-law behavior can be explained by Eq. (2), in which the integral becomes a constant at $H = H_s$, where $\mu = 0$, yielding

$$M_s - M = 0.241\,32 g\mu_B \sqrt{k_B T/J} \qquad (4)$$

per Cu^{2+}, because $m = \hbar^2/J$. As shown in Fig. 3(b), the equation $M_s - M = B\sqrt{k_B T/J}$ can be fitted very well to the 14 T data over the entire temperature range of the measurements, up to 15 K, by choosing M_s and B as separate fitting parameters, while J is set at 10.8 K obtained from the $M(H)$ data. The fit gives $M_s = 1.14\mu_B$ per Cu^{2+}, in excellent agreement with 1.15 μ_B obtained from the $M(H)$ data (see Fig. 1), and $B = 0.230(1)\ g\mu_B$ in good agreement with the exact prefactor in Eq. (4). Moreover, as is also shown in the figure, the data are in nearly perfect agreement with our QTM calculation at H_s using the J and g obtained from the $M(H)$ data.

At this field, specific heat divided by temperature, shown in the inset to Fig. 3(b), also exhibits characteristic power-law behavior. The best fit of the relation $C/T \propto T^{-\alpha}$ to the data below 2 K yields $\alpha = 0.49(1) \approx 1/2$. This power-law dependence arises directly from the density of states in one

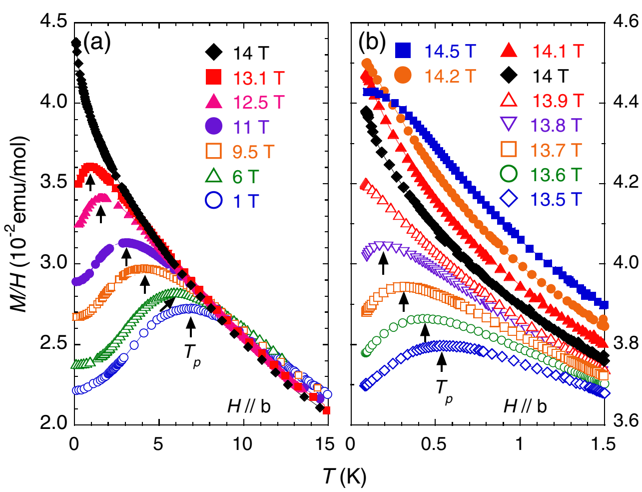

FIG. 2 (color online). (a) Temperature dependence of M/H at various fields. The black arrows indicate the peak position T_p. (b) Low-temperature part ($T \le 1.5$ K) of the M/H plot for magnetic fields slightly below (open symbols) and slightly above (solid symbols) H_s. Thin lines are guides to the eye.

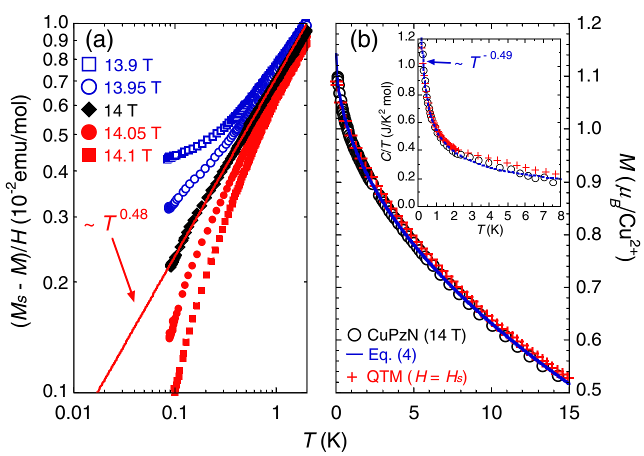

FIG. 3 (color online). (a) Log-log plot of $(M_s - M)/H$ near H_s as a function of temperature below 2 K. The saturation magnetization $M_s = 1.15\mu_B$ has been taken from the magnetization curve at 0.08 K (see Fig. 3). The best fit of a power law, $(M_s - M)/H \propto T^{\beta}$, to the 14 T data yields $\beta = 0.48(1)$ (solid line). (b) Comparison of M at 14 T with the result of a QTM calculation for a 1D spin-1/2 HAF at H_s (crosses). The solid line is the best fit with $\beta = 1/2$ described in the text. Inset: C/T as a function of temperature at 14 T (open circles). Nuclear and phonon contributions have been subtracted. Crosses are QTM results. The best fit of the power law $C/T \propto T^{-\alpha}$ below 2 K yields $\alpha = 0.49(1)$ (dotted line).

dimension, $D(\epsilon) \propto 1/\sqrt{\epsilon}$: since C/T is approximately proportional to $D(k_B T)$ when $\mu = 0$, it follows that it is proportional to $1/\sqrt{T}$ [34]. To be precise,

$$C/T = 0.22894 k_B^{3/2}/\sqrt{JT} \quad (5)$$

per Cu^{2+}. Fitting the expression $C/T = A/\sqrt{JT}$ to the data below 2 K—where A is the only fitting parameter, with J the one obtained from the $M(H)$ data—yields $A = 0.215(1)k_B^{3/2}$ in good agreement with the exact prefactor in Eq. (5). As is also shown in the figure, the data are in excellent agreement with our QTM calculation using the J obtained from the $M(H)$ data.

At fields slightly away from H_s, the $(M_s - M)/H$ vs T plots in Fig. 3(a) deviate from the $\sqrt{T}$ behavior at low temperatures but retain it above 1 K. This trend can also be explained by Eq. (2). Since H and T appear in the integrand of Eq. (2) only as the combination $\mu/k_B T$, Eq. (4) holds for $k_B T \gg g\mu_B(H_s - H)$ as long as the dispersion is quadratic. It should be emphasized, however, that the $\sqrt{T}$ behavior persists down to $T = 0$ only at H_s.

A brief remark on the power-law exponents is in order. Obviously, the combination $\alpha + \beta(1 + \delta) = 1.92(4)$ of the exponents $\alpha = 0.49(1)$, $\beta = 0.48(1)$, and $\delta = 1.98(8)$ from our experiment is very close to the universal scaling value 2. In fact, $\alpha = 1/2$ can be obtained simply from the scaling relation $\alpha = 2 - (d + z)/z$, where the dynamical exponent z is 2 for free fermions and the spatial dimension d is 1. Similarly, $\beta = 1/2$ and $\delta = 2$ can be derived by employing a scaling argument [9].

Finally, the magnetic phase diagram of CuPzN is presented in Fig. 4 on the basis of $d(M/H)/dT$, with T_p from Fig. 2 superposed to indicate the crossover to the TLL phase. Note that Eq. (3) gives a parameter-free expression for T_p,

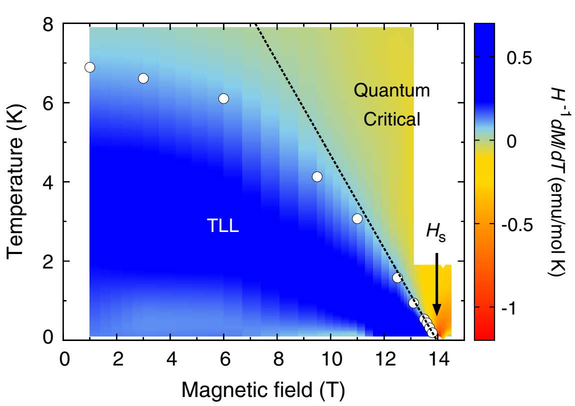

FIG. 4 (color online). T vs H phase diagram of CuPzN based on the temperature derivative of M/H. Open circles denote the positions of T_p, the temperature of the broad peak in M. The dotted line is the universal crossover line for the free-fermion limit, Eq. (6).

$$T_p = 0.76238 \frac{g\mu_B}{k_B}(H_s - H). \quad (6)$$

This universal relation, shown as a dotted line with the g and H_s obtained from the $M(H)$ data, with no fitting parameter, agrees excellently with the data near H_s. The linear dependence, distinct from the power-law dependence for a Bose-Einstein condensation (BEC) of magnons [35], indicates that the 3D magnetic ordering of CuPzN due to J' is irrelevant in the temperature range of the present work, at least near H_s. This is further supported by the 1D exponents for the specific heat and magnetization, $\alpha = 0.49(1) \approx 1/2$ and $\beta = 0.48(1) \approx 1/2$, which are in marked contrast to $\alpha = -1/2$ and $\beta = 3/2$ found in the magnon BEC in $NiCl_2$-$4SC(NH_2)_2$ [36]. As the magnetic field further decreases, T_p deviates downward from the straight line, owing to repulsion between magnons [6].

In summary, we have examined in detail a crossover of CuPzN from a thermally disordered high-temperature phase to the Tomonaga-Luttinger-liquid phase, and the critical behavior of the magnetization and specific heat near the saturation field H_s. The crossover temperature T_p—the temperature of the broad magnetization peak—starts off at a low field with the theoretical value that has been well known for 50 years [29] for the one-dimensional spin-1/2 Heisenberg model, decreases with increasing field, and smoothly connects near H_s to the universal, linear line for free fermions. At H_s, the magnetization and specific heat are in excellent agreement with universal power laws for free fermions and with exact results calculated with the QTM method. The magnetization curve at 0.08 K is also in excellent quantitative agreement with an exact Bethe-ansatz result up to 99% of H_s. The deviation very near H_s is fully accounted for by QTM calculations at this temperature. These findings demonstrate that CuPzN is a practically perfect one-dimensional spin-1/2 Heisenberg antiferromagnet.

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